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Citation for final published version:

Mo, Liwu, Zhang, Feng, Deng, Min, Jin, Fei ORCID: <https://orcid.org/0000-0003-0899-7063>, Al-Tabbaa, Abir and Wang, Aiguo 2017. Accelerated carbonation and performance of concrete made with steel slag as binding materials and aggregates. Cement and Concrete Composites 83 , pp. 138-145. 10.1016/j.cemconcomp.2017.07.018 file

Publishers page: <http://dx.doi.org/10.1016/j.cemconcomp.2017.07.018>  
<<http://dx.doi.org/10.1016/j.cemconcomp.2017.07.018>>

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# Accelerated carbonation and performance of concrete made with steel slag as binding materials and aggregates



Liwu Mo<sup>a,\*</sup>, Feng Zhang<sup>a</sup>, Min Deng<sup>a,b</sup>, Fei Jin<sup>c</sup>, Abir Al-Tabbaa<sup>c</sup>, Aiguo Wang<sup>d</sup>

<sup>a</sup> College of Materials Science and Engineering, Nanjing Tech University, Nanjing, 210009, China

<sup>b</sup> State Key Laboratory of Materials-Oriented Chemical Engineering, Nanjing, 210009, China

<sup>c</sup> Department of Engineering, University of Cambridge, CB2 1PZ, UK

<sup>d</sup> Anhui Key Laboratory of Advanced Building Materials, Anhui Jianzhu University, Hefei, 230022, China

## ARTICLE INFO

### Article history:

Received 19 February 2017

Received in revised form

16 June 2017

Accepted 18 July 2017

Available online 23 July 2017

### Keywords:

Carbonation

Steel slag

Compressive strength

Microstructure

Expansion

## ABSTRACT

Steel slag has been used as supplementary cementitious materials or aggregates in concrete. However, the substitution levels of steel slag for Portland cement or natural aggregates were limited due to its low hydraulic property or latent volume instability. In this study, 60% of steel slag powders containing high free-CaO content, 20% of Portland cement and up to 20% of reactive magnesia and lime were mixed to prepare the binding blends. The binding blends were then used to cast concrete, in which up to 100% of natural aggregates (limestone and river sands) were replaced with steel slag aggregates. The concrete was exposed to carbonation curing with a concentration of 99.9% CO<sub>2</sub> and a pressure of 0.10 MPa for different durations (1d, 3d, and 14d). The carbonation front, carbonate products, compressive strength, microstructure, and volume stability of the concrete were investigated. Results show that the compressive strength of the steel slag concrete after CO<sub>2</sub> curing was significantly increased. The compressive strengths of concrete subjected to CO<sub>2</sub> curing for 14d were up to five-fold greater than that of the corresponding concrete under conventional moist curing for 28d. This is attributed to the formation of calcium carbonates, leading to a microstructure densification of the concrete. Replacement of limestone and sand aggregates with steel slag aggregates also increased the compressive strengths of the concrete subjected to CO<sub>2</sub> curing. In addition, the concrete pre-exposed to CO<sub>2</sub> curing produced less expansion than the concrete pre-exposed to moist curing during the subsequent accelerated curing in 60 °C water. This study provides a potential approach to prepare concrete with low-carbon emissions via the accelerated carbonation of steel slag.

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## 1. Introduction

Manufacturing of Portland cement is an intensively energy-consuming and CO<sub>2</sub>-emitting process, which accounts for around 5%–8% of the total man-made CO<sub>2</sub> emissions globally [1]. On one hand, to produce low-carbon alternative cements is of great significance for the prospect of developing sustainable construction materials. On the other hand, CO<sub>2</sub> curing for construction materials has recently gained increasing attention owing to its potential of reducing the carbon emissions [2,3]. This CO<sub>2</sub> curing technology provides a new way for integrating the economical, permanent, and safe sequestration of CO<sub>2</sub> and producing low-carbon construction materials. In some previous studies, alternative binders based on calcium carbonate were developed via carbonating CaO-rich raw

materials [4,5]. In addition, reactive magnesia was also used to prepare magnesium carbonate binders [3]. The presence of MgO enhanced the carbonation of Ca(OH)<sub>2</sub> and improved the mechanical strength accordingly [5]. Our previous studies found that Mg<sup>2+</sup> could change the morphology of CaCO<sub>3</sub> and make the carbonate products pack more closely, facilitating the mechanical strength improvement [6,7]. As the manufacturing of MgO and CaO also produce CO<sub>2</sub>, seeking for alternative more economical CaO and MgO resources for preparing the carbonated binders would be of great interest and significance.

Steel slag is a type of byproduct produced during the steel manufacturing. In 2015, 120 million tonnes of steel slag was produced in China with another >1 billion tonnes accumulated, while the total utilization ratio is only 10%. Most of the steel slag was occupying a huge amount of farmland and causing environmental pollution of the soils and underground water. Steel slag contains

\* Corresponding author.

E-mail address: [andymoliwu@njtech.edu.cn](mailto:andymoliwu@njtech.edu.cn) (L. Mo).

high contents of CaO as well as MgO. The utilization of steel slag as construction materials, e.g. supplementary cementitious materials, aggregates, etc. has been widely studied [8–12]. However, steel slag exhibits poor hydraulic property due to its lack of tri-calcium silicates and amorphous SiO<sub>2</sub> content [10,13]. Therefore, high replacement of Portland cement with the steel slag resulted in the decrease of mechanical strength development [14]. In addition, steel slag containing high content of free-MgO/CaO may cause excessive expansion and hence induce volume instability of the materials in the long term, leading to a limited replacement level as a cementitious material. Steel slag was also used as aggregates in concrete [11,12], but the risk of excessive expansion exists if it contains high contents of free-MgO/CaO.

Nonetheless, the high contents of free-MgO/CaO in the steel slag make it a potential candidate for carbonated binder and it has already been proved to have high carbonation reactivity [15–19]. For example, compacted steel slag paste with high compressive strength of up to 80 MPa was prepared by CO<sub>2</sub> curing [15]. This is attributed to the microstructure densification of the materials as a result of formation of calcite (CaCO<sub>3</sub>). Moreover, carbonation of the expansive components (mainly free-MgO/CaO) in the steel slag was reported to effectively improve the soundness of mortars using steel slag as partial replacement for natural aggregates [20]. However, so far there have been very limited studies on the incorporation of high volume of steel slag as both binding materials and aggregates in concrete under CO<sub>2</sub> curing. The aim of the present study is to investigate the possibility of utilizing steel slag at a high replacement level (60% in binding materials and 100% in aggregates) to produce concretes using the accelerated carbonation technology. Reactive magnesia and lime were also used to prepare the binding materials with different MgO and CaO contents, considering more calcium and magnesium sources are available for the formation of carbonates. To facilitate the penetration and diffuse of CO<sub>2</sub> into the concrete specimens, CO<sub>2</sub> with a high concentration of 99.9% and a pressure of 0.1 MPa was used for carbonation. The effects of 99.9% CO<sub>2</sub> curing under the pressure of 0.1 MPa on the carbonation front, compressive strengths, microstructure, and volume stability of the concretes were thoroughly investigated and discussed. This study may provide a potential approach to make steel slag concrete with high replacement levels, and also to integrate the construction materials manufacturing with CO<sub>2</sub> storage.

## 2. Materials and experimental methods

### 2.1. Raw materials

Portland cement (type P.II 52.5) manufactured by the Onoda Cement Co., Ltd., Nanjing was used. Converter slag was provided by Baosteel Co., Ltd., Shanghai, which was produced in a Basic Oxygen Furnace (BOF). Quicklime and reactive magnesia were produced by calcining limestone and magnesite, respectively. Table 1 shows the chemical compositions of the Portland cement, lime, reactive magnesia, and steel slag. According to the XRD patterns of the steel slag shown in Fig. 1, the steel slag mainly consists of various types of calcium silicate and Portlandite (hydration products of free-CaO). The MgO exists mainly in the forms of periclase and solid solutions, while the CaO exists mainly in the forms of calcium silicate, free-CaO, Portlandite, and other solid solutions. It should be noted that content of free-CaO is 8.38%. Steel slag was ground into particles with different size ranges. Steel slag powders with particle size smaller than 75 μm was used as a replacement of Portland cement. Steel slag with particle size smaller than 5 mm was used as fine aggregates, and coarse steel slag with size of 5–16 mm was used directly as the coarse aggregates for concrete. Fig. 2 shows the

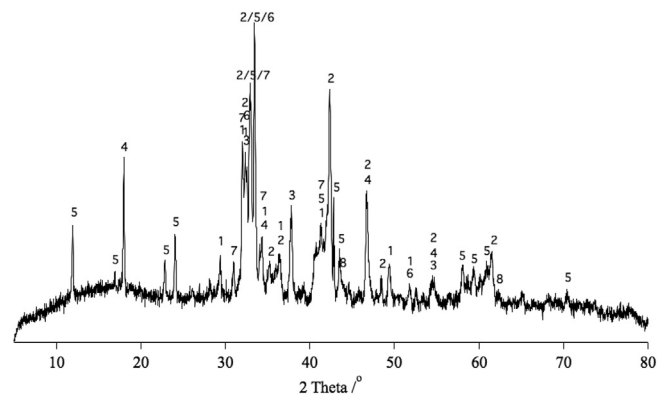
**Table 1**

Chemical compositions of Portland cement, lime, reactive magnesia and steel slag/wt.%.

	Cement	Lime	Reactive MgO	Steel slag
MgO	2.35	13.39	88.52	7.19
CaO	64	82.19	3.18	42.42 (8.38 <sup>b</sup> )
SiO <sub>2</sub>	19.43	1.63	2.76	11.04
Fe <sub>2</sub> O <sub>3</sub>	2.96	0.11	0.26	27.37
Al <sub>2</sub> O <sub>3</sub>	4.73	0.13	0.42	1.61
SO <sub>3</sub>	2.58	0.04	0.61	0.11
K <sub>2</sub> O	0.68	0.06	0.03	—
Na <sub>2</sub> O	0.11	0.05	0.06	—
MnO	—	—	—	2.28
P <sub>2</sub> O <sub>5</sub>	—	—	—	2.09
TiO <sub>2</sub>	—	—	—	0.5
Cr <sub>2</sub> O <sub>3</sub>	—	—	—	0.15
V <sub>2</sub> O <sub>5</sub>	—	—	—	0.13
IL <sup>a</sup>	2.81	2.15	3.62	—

<sup>a</sup> Ignition loss.

<sup>b</sup> f-CaO.



**Fig. 1.** The XRD pattern of steel slag (1: Calcium silicate (Ca<sub>3</sub>SiO<sub>5</sub>), 2: Beta-Calcium silicate (Ca<sub>2</sub>SiO<sub>4</sub>), 3: Lime, 4: Portlandite, 5: Brownmillerite, 6: alpha-Calcium Silicate (Ca<sub>2</sub>SiO<sub>4</sub>), 7: beta-Larnite, 8: Periclase).

optical images of steel slag powders and aggregates. For comparison, limestone and river sands were used as the natural aggregates for concrete.

### 2.2. Preparation and CO<sub>2</sub> curing of concrete

Binding materials were firstly prepared by mixing steel slag powders, Portland cement, magnesia, and quicklime according to the formulation of binding materials shown in Table 2. Accordingly, two types of binding materials were prepared, which were designated as “SCM” and “SCLM” respectively. SCM represents that the binding materials consist of 60% of steel slag, 20% of Portland cement, and 20% of reactive magnesia, while in SCLM 10% of quicklime was incorporated in the binding materials as a replacement of reactive magnesia. Letters “N” and “S” indicate that the concrete were cast with natural aggregates (limestone as coarse aggregates, and sand as fine aggregates) and with steel slag (both coarse and fine aggregates) respectively.

Concrete specimens were cast with the same mix proportion of 340 kg/m<sup>3</sup> binding materials, 1200 kg/m<sup>3</sup> coarse aggregates, 675 kg/m<sup>3</sup> fine aggregates, and 185 kg/m<sup>3</sup> water (water to binder ratio of 0.54). For each mix proportion, twelve cubic concrete specimens with size of 100 × 100 × 100 mm were cast. The cubic specimens were used to examine the compressive strength. After being cast, all the concrete specimens were cured in a moist room with 98% relative humidity and temperature of 23 °C for 24 ± 2 h, and then demoulded. Thereafter, the concrete cubes were cured in





Fig. 2. Optical images of steel slag powders and aggregates: (a) steel slag powders; (b) and (c) <5 mm fine slag aggregates; (d) 5–16 mm coarse slag aggregates.

**Table 2**  
Mix proportions of concrete/wt.%.

Mix design	Binding materials				Fine aggregates	Coarse aggregates
	Steel slag	Portland cement	Reactive magnesia	Lime		
SCM-S	60	20	20	0	Steel slag	Steel slag
SCLM-S	60	20	10	10	Steel slag	Steel slag
SCM-N	60	20	20	0	Sands	Limestone
SCLM-N	60	20	10	10	Sands	Limestone

the moist room for 7 more days and then exposed to CO<sub>2</sub> curing with a concentration of 99.9% and a pressure of 0.10 MPa for different durations of 1, 3, and 14 d respectively. For comparison, six concrete cubes for each mix proportion were cast and cured in the moist room until the compressive strength test at 7 and 28 d.

In order to evaluate the volume stability, prismatic concrete specimens with size of 100 × 100 × 400 mm for each mix proportion were prepared. Similar to the cubic concrete specimens, the prismatic concrete specimens were subjected to CO<sub>2</sub> curing for 14 d after the 7 d of moist curing. Subsequently, the concrete prisms were immersed in water at 60 °C to accelerate the hydration of periclase and free lime that might have not been carbonated or hydrated. A strain gauge was embedded into the central part of the concrete prism horizontally during the casting process. The deformation of concrete specimens was recorded automatically with a computer, and was zeroed at 3 h after being immersed into the 60 °C water for excluding the thermal expansion due to the temperature rise. For comparison, concrete prisms that were continuously placed in the moist room for 14 d, and then were also cured in the 60 °C water to investigate their linear deformation.

### 2.3. Performance test procedure

At each curing age as mentioned above, the carbonation depth of the concrete was examined by spraying phenolphthalein indicator and compressive strengths of concrete were tested. For the compressive strength test at each age, the mean value of three

specimens was used.

Mortar sample in the concrete was extracted via carefully removing the coarse aggregates first. Then the mortar sample was crushed and ground gently into powders with a pestle and mortar, and during this process the fine aggregates were tweezed once they were observed. The powder sample was passed through a 224 μm sieve. The fraction passed through the 224 μm sieve was ground further with the mortar and pestle until the entire sample passed through the 75 μm sieve while the fraction retained on the 224 μm sieve was removed considering it usually contained high content of fine aggregates. The powders with particle sizes less than 75 μm were used to examine the carbonation products formed in concrete during CO<sub>2</sub> curing by using X-ray diffraction (XRD, SmartLab X-ray diffractometer, Rigaku Co. Japan) with Cu Kα radiation ( $\lambda = 1.5418 \text{ \AA}$ ) and a scanning  $2\theta$  range of 5°–80°. Slices with thickness of 5 mm were sawn from the carbonated concrete specimens, and then vacuum dried at 60 °C for 24 ± 2 h. The dried slices were epoxy impregnated, polished, carbon coated, and then investigated by Backscattered Electronic microscopy (BSE) coupled with Energy Dispersive X-ray (EDX), using JEOL JSM-5900LV SEM to characterize the microstructure.

## 3. Results and discussion

### 3.1. Carbonation front

Fig. 3 shows the optical images of the concrete specimens after

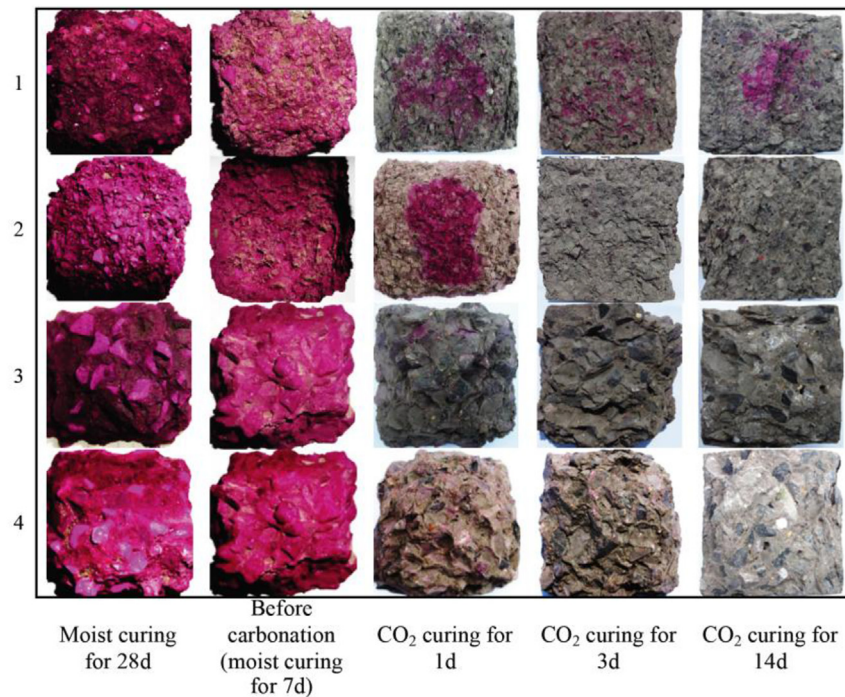


Fig. 3. Optical images of concrete specimens sprayed with phenolphthalein indicator (1: SCM-S, 2: SCLM-S, 3: SCM-N, 4: SCLM-N).

being sprayed with phenolphthalein indicator. All the concrete specimens cured under moist condition showed obvious pink across the whole section, whereas after being exposed to  $\text{CO}_2$  the pink area was reduced and gradually disappeared with the increase of exposure time to 14d. This indicates that the  $\text{CO}_2$  has gradually penetrated throughout the whole sections of specimens and reacted with the relevant MgO and CaO-bearing phases, e.g. Portlandite, calcium silicate, magnesia, lime, etc. It is also clear that the SCM-S sample is the most resistant to  $\text{CO}_2$  penetration since even after 14 d of accelerated carbonation, there is still a portion of the core remaining uncarbonated. This could be ascribed to the denser

structure formed before and during the accelerated carbonation process, which will be discussed below.

Fig. 4 shows the XRD patterns of the mortar samples extracted from concrete specimens. Accordingly, for the concrete without  $\text{CO}_2$  curing, peaks of the most notable hydration products, i.e., Portlandite and brucite were observed. Portlandite was formed due to the hydration of Portland cement, free-CaO contained in steel slag, and the added quicklime. Brucite was the hydration product of the reactive MgO. For the concrete with  $\text{CO}_2$  curing, calcite was the main carbonate product by the consumption of Portlandite and many other Ca-rich minerals such as brownmillerite and various forms of calcium silicates contained in Portland cement and steel slag. This was confirmed by the disappearance of Portlandite peaks and relatively weaker peaks of calcium silicates (particularly alpha- and beta-calcium silicate) identified in the carbonated samples. Meanwhile, the diffraction peaks of brucite were absent in the mortar samples after carbonation, which may be ascribed to its transformation to the various hydrated magnesium carbonates [3,5–7]. However, due to the limited amount of MgO in the mortar samples, these Mg-carbonates were not identified from the XRD patterns. In addition, magnesium could also be incorporated in the calcite structure during carbonation resulting in Mg-bearing calcite as indicated by previous studies [6]. From the XRD analysis, no obvious influence was observed by the incorporation of quicklime on the types of hydration or carbonation products but it may affect their quantities.

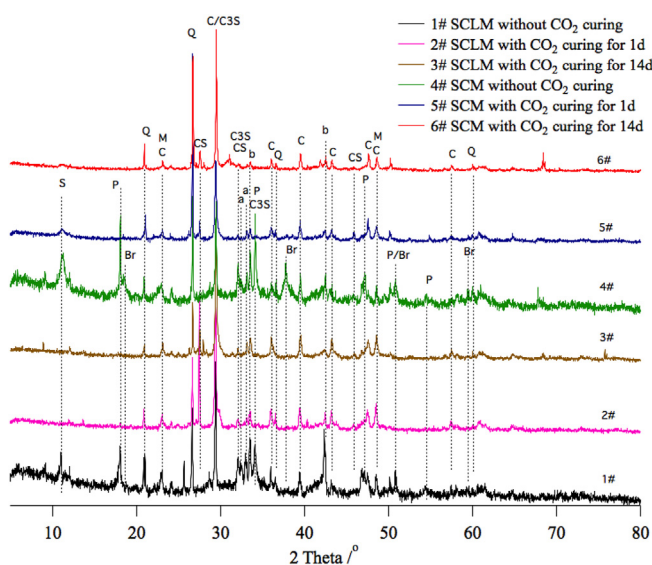


Fig. 4. XRD patterns of mortar samples SCM-S and SCLM-s extracted from concrete exposed to moist curing and  $\text{CO}_2$  curing (a: Alpha-Calcium silicate ( $\text{Ca}_2\text{SiO}_4$ ), b: Beta-Calcium Silicate ( $\text{Ca}_2\text{SiO}_4$ ), Br: Brucite, CS: Wollastonite ( $\text{CaSiO}_3$ ), C<sub>3</sub>S: Alite ( $\text{Ca}_3\text{SiO}_5$ ), C: Calcite, P: Portlandite, Q: Quartz, M: Brownmillerite).

### 3.2. Compressive strength

Table 3 shows the compressive strengths of concrete specimens cured under moist condition and exposed to  $\text{CO}_2$  curing. All the concrete specimens cured under moist curing condition exhibited relatively low compressive strengths. For example, the compressive strengths of concrete specimens at 7 d ranged from 7.9 to 10.7 MPa. With the curing age increasing to 28 d under moist curing, the compressive strengths increased slightly regardless of the type of



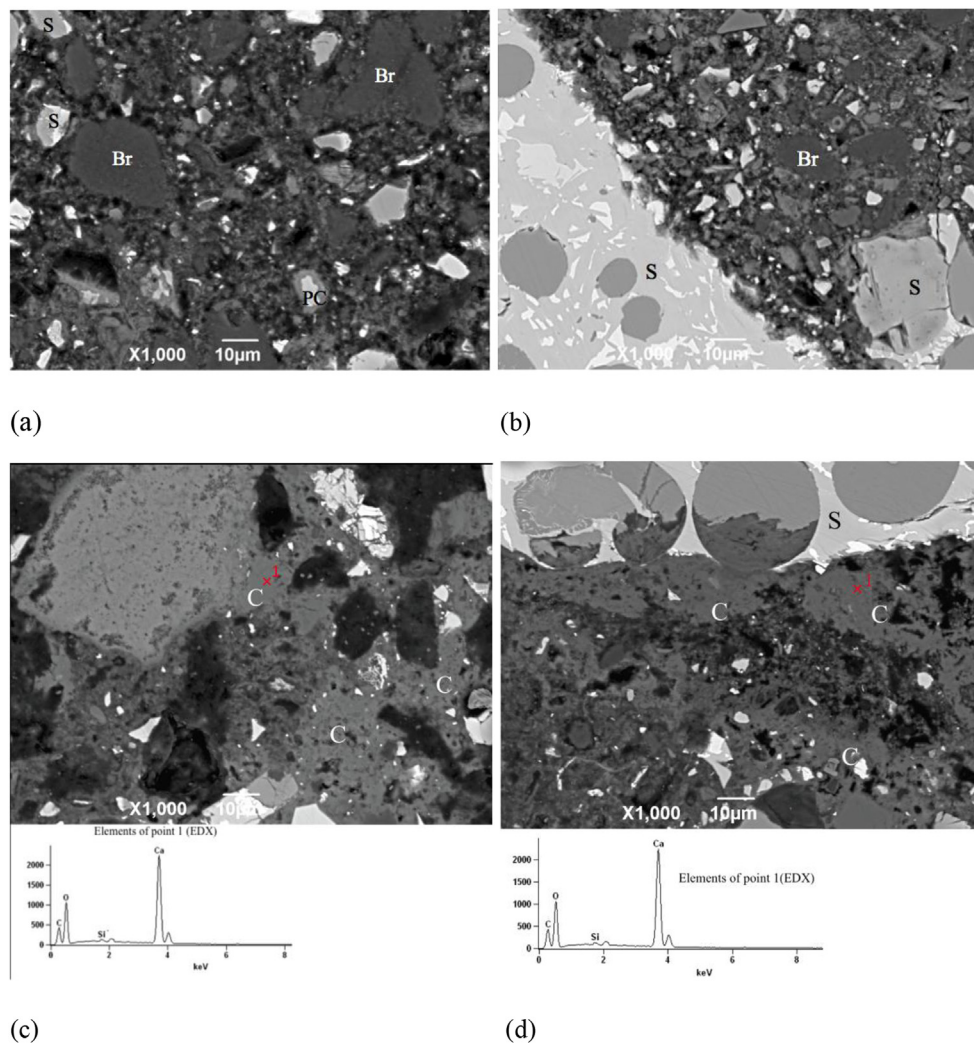
**Table 3**Compressive strength of concrete specimens subjected to moist curing and CO<sub>2</sub> curing/MPa.

Mix design	Moist curing	CO <sub>2</sub> curing			
	28d	Before carbonation (moist cured for 7d)	1d	3d	14d
SCM-S	11.5	10.7	38.6	52.7	61.3
SCLM-S	10.5	9.4	33.5	48.4	55.9
SCM-N	10.7	9.0	24.6	40.8	46.2
SCLM-N	9.4	7.9	27.6	36.6	41.2

coarse aggregates, which is due to the large replacements of the Portland cement with low hydraulic steel slag. In contrast, after exposure to CO<sub>2</sub> curing for only 1 d, the compressive strengths of all the concrete specimens were significantly increased. For the concrete made with natural limestone aggregates after only 1 d of CO<sub>2</sub> curing, in comparison to the concrete prior to the CO<sub>2</sub> exposure (namely after 7 d of moist curing), the compressive strengths were increased by 2.7–3.5 times, while those of the concrete made with steel slag as fine and coarse aggregates were increased by approximately 3.6–3.8 times.

Moreover, with increasing CO<sub>2</sub> exposure time, the compressive strengths were increased dramatically. After 14 d of CO<sub>2</sub> curing, the compressive strengths of concrete SCM-N, SCM-S, SCLM-N, and

SCLM-S were 46.2 MPa, 61.3 MPa, 41.2 MPa, and 55.9 MPa respectively, which were 4.2–9.5 folds higher than those of the concrete specimens cured under moist condition for 7 d, and were 4.3–5.3 times higher than those of the concrete specimens after 28 d of moist curing. This is attributed to the carbonation reaction of the Mg- and Ca-rich minerals in the concrete specimens during CO<sub>2</sub> curing, which produced various strength-contributing carbonate products as confirmed by many other studies [3,6,7]. It is also clear that the strength enhancement rate was reduced with CO<sub>2</sub> curing time due to (a) the fast carbonation reaction in the first few days, which would be the case for samples SCM-N, SCLM-N and SCMS-S or (b) the denser microstructure formed during the carbonation process leading to more difficult CO<sub>2</sub> penetration in the samples,



**Fig. 5.** Typical BSE images of the concrete subjected to moist curing and CO<sub>2</sub> curing: (a) mortar matrix in concrete SCM-S subjected to moist curing, (b) interface between steel slag aggregates and matrix in concrete SCM-S subjected to moist curing, (c) mortar matrix in concrete SCM-S subjected to CO<sub>2</sub> curing, (d) interface between steel slag aggregates and matrix in concrete SCM-S subjected to CO<sub>2</sub> curing. Br: Brucite, C: Calcite, S: Steel slag.

which corresponds to the case of sample SCM-S (see Fig. 3). In addition, regardless of the aggregates type, prolonged carbonation time caused more augments in compressive strength of the concrete containing higher content of reactive magnesia. In comparison to the concrete cast with natural limestone and sands, the concrete prepared with steel slag aggregates exhibited higher compressive strengths due to their higher amounts of minerals that could be carbonated.

### 3.3. Microstructure

Fig. 5 shows the typical BSE images of concrete containing steel slag aggregates subjected to moist curing and CO<sub>2</sub> curing. Under moist curing condition, the microstructure of mortar matrix in concrete was porous (Fig. 5a). This is understandable as only 20% of Portland cement was used, which produced a small amount of hydration products in the concrete. In addition, a porous interface between the coarse steel slag aggregate and the mortar matrix was also observed in Fig. 5b. In comparison, as shown in Fig. 5c, the mortar matrix in the carbonated concrete was much denser, owing to the formation of calcite as a result of carbonation. Moreover, a denser interface between the coarse steel slag aggregate and mortar matrix were observed in the carbonated concrete (Fig. 5d).

### 3.4. Volume deformation under accelerated curing in 60 °C water

Fig. 6 shows the expansion curves of concrete prisms with and without accelerated carbonation followed by curing in 60 °C water. Accordingly, all the concrete prisms without carbonation treatment produced great expansions when cured in the 60 °C water. For example, the expansions of SCM-N and SCLM-N without CO<sub>2</sub> treatment reached 500 and 250 microstrains at 60 d respectively. The expansion was mainly caused by not only the hydration of periclase and free-CaO existing in the steel slag but also the lime and magnesia added. The higher the incorporation of magnesia, the larger the expansion was produced due to the larger expansion of MgO compared to CaO upon hydration. In comparison to the concrete prisms cast by using the natural limestone and river sand aggregates, the concrete prisms prepared with steel slag as fine and coarse aggregates produced much higher expansion, and even cracked the specimens (as shown in Fig. 7a and c), which implies that the hydration of periclase and free-CaO contained in the steel slag aggregates contributed significantly to the expansion. SCM-S and SCLM-S reached maximum expansions of 4500 and 4000 microstrain around 55 d in the 60 °C water. After then the deformation curves of both SCM-S and SCLM-S declined. This is corresponding to deterioration of concrete caused by cracking (Fig. 7a and (c)), which could not force the strain gauge to produce equivalent expansion simultaneously. Regardless of the aggregates type, the CO<sub>2</sub> treatment on the concrete prisms dramatically reduced the expansion. This can be attributed to the fact that the contents of periclase and free-CaO in the steel slag, quicklime and reactive magnesia added had been significantly reduced due to carbonation as shown in the XRD analysis (Fig. 4). Fig. 7 shows the images of the concrete specimens after 60 d curing in the 60 °C water. As shown in Fig. 7, the concrete without carbonation treatment were deteriorated owing to its excessive volume expansion, while good appearance of the concrete with previous CO<sub>2</sub> curing was observed, agreeing well with the deformation test.

## 4. Discussion

Under conventional moist curing, the steel slag powders exhibited low binding properties and contributed little to the

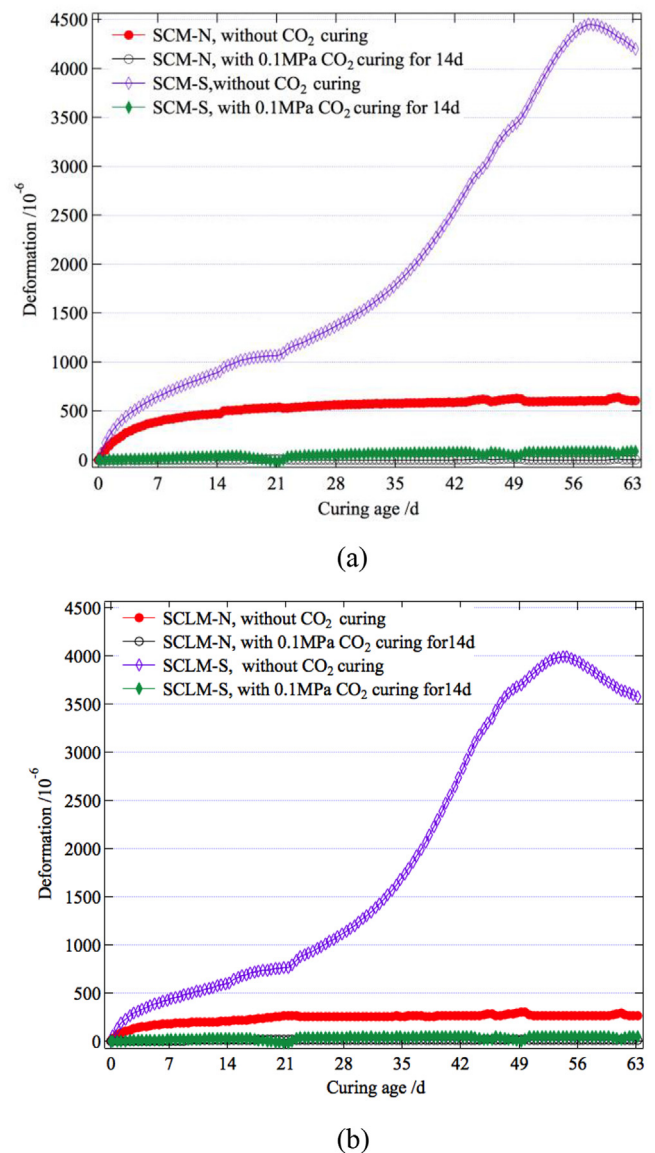


Fig. 6. Expansion curves of the concrete specimens after being exposed to moist curing or CO<sub>2</sub> curing and then followed by curing in 60 °C water: (a) concrete made with SCM, and (b) concrete made with SCLM.

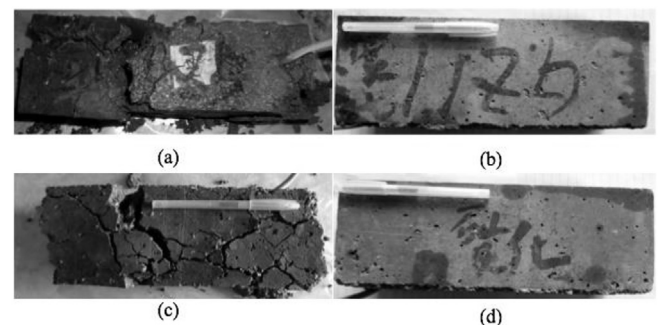


Fig. 7. Optical images of the concrete specimens after being exposed to moist curing or CO<sub>2</sub> curing and then followed by curing in 60 °C water for 60 d: (a) SCM-S, moist curing previously, (b) SCM-S, CO<sub>2</sub> curing previously, (c) SCLM-S, moist curing previously, and (d) SCLM-S, CO<sub>2</sub> curing previously.

compressive strength of concrete. This is due to the lack of minerals with relatively high hydraulic activity in the steel slag [10,13,14]. However, under carbonation condition with a  $\text{CO}_2$  pressure of 0.1 MPa, the  $\text{CO}_2$  could penetrate through the whole concrete specimens rapidly to react with the carbonation components in the steel slag, e.g. calcium silicates, free-CaO, Portlandite, etc., and hence formed calcium carbonate (e.g., calcite), generating high binding performance, which agrees with previous studies [15,16]. The compressive strength enhancement of the steel slag concrete with  $\text{CO}_2$  curing is attributed to the microstructure densification of concrete owing to the formation of carbonate products. Apart from the minerals in the steel slag, the hydration products of Portland cement, unhydrated Portland cement, reactive magnesia, and quicklime could also be carbonated and contributed to the formation of these strength-giving carbonates. Addition of quicklime in the blends was expected to provide more calcium content for the calcite formation but it was found that it actually decreased the compressive strength of concrete in comparison to the SCM blends. This implies that the carbonation products of magnesia (either hydrated magnesium carbonates [3] or Mg-bearing calcite [6,7]) contributes more to the strength enhancement than the pure calcite. The most obvious evidence can be referred to Fig. 3, where the partially carbonated SCM-S gives higher strength than the fully carbonated SCLM-S at 14 d, showing that although the replacement of magnesia by quicklime enhanced the carbonation degree, it leads to more porous matrix and decreases the overall strength.

Incorporation of steel slag in concrete as fine and coarse aggregates caused no obvious influences on the compressive strength of concrete under moist curing, but dramatically increased the compressive strength of the concrete when subjected to  $\text{CO}_2$  curing (Table 3). This is related to interface enhancement due to the formation of calcite in the interface zone, densifying the interface microstructures (Fig. 5d). The full replacement of the nature aggregates also resulted in much lower carbon footprint of the concrete and significantly improved the sustainability of these construction materials.

The deformation examination on the carbonated concrete during the continuous curing in  $60^\circ\text{C}$  water indicates that the volume of carbonated steel slag concrete is stable within the test age in comparison to the moist cured concrete. This is related to the contents of expansive free-CaO and periclase intrinsically contained in steel slag, and that from the added quicklime and magnesia, all of which were notably reduced due to the carbonation

reaction. While for the moist cured concrete, the continuous hydration of unhydrated MgO and CaO was accelerated and produced excessive expansion during the subsequent curing in the  $60^\circ\text{C}$  water, eventually cracking the concrete. In the moist cured concrete, cracked coarse steel slag aggregate was observed as shown in Fig. 8, which is due to the hydration of free-CaO contained in the coarse steel slag aggregate. Under  $\text{CO}_2$  curing, dense carbonate products formed around the coarse steel slag particle act as a protection layer, preventing the water from diffusing into the inner part of the steel slag aggregate, and therefore inhibit continuous hydration of the expansive components of free-CaO and periclase. This significantly improves the volume stability of steel slag concrete. This study demonstrated that the  $\text{CO}_2$  treatment could be a potential approach to utilize the steel slag as both binder and aggregates in concrete. Nevertheless, the long term volume stability of the steel slag concrete needs to be investigated further.

The carbonation approach developed in this study seems not to be suitable for standard in-situ use in constructions, however it could be used for precast products manufacturing in a factory, e.g. concrete blocks, pervious concrete, bricks, etc. Further study on application of the carbonation technology is needed, which would be of great interest and significance.

## 5. Conclusions

The effects of  $\text{CO}_2$  curing on the carbonation depth, compressive strength, and volume stability of the concrete made with steel slag as binding materials and aggregates were investigated. Some conclusions can be drawn as follows:

- 1 Based on the phenolphthalein indicator test, with 0.1 MPa  $\text{CO}_2$  curing, the concrete specimens were almost carbonated within 3d, indicating it is effective to accelerate the carbonation of concrete via pressurized  $\text{CO}_2$ .
- 2 Under conventional moist curing, the compressive strengths of concrete cast with the binding blends consisting of 60% steel slag powders, up to 20% of lime and magnesia, and only 20% of Portland cement were very low, being 7.9–10.7 MPa at 7 d and 9.4–11.5 MPa at 28 d. However, they were increased dramatically during the following  $\text{CO}_2$  curing. The compressive strength of concrete subjected to  $\text{CO}_2$  curing for 14 d were increased by 4.3–5.3 folds in comparison to those of the concrete moist cured

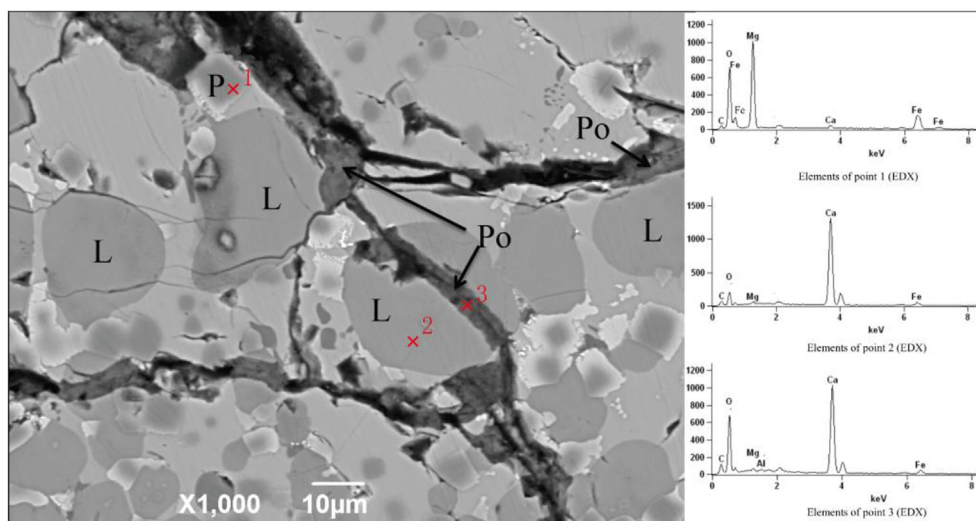


Fig. 8. BSE image of a cracked steel slag aggregate in moist cured steel slag concrete (L: free-CaO, P: Periclase, Po: Portlandite).



for 28 d. This is related to the densification of concrete caused by the formation of carbonate products.

- 3 Under moist curing, replacement of natural aggregates (lime stone and river sand) with steel slag caused negligible influences on the compressive strength of concrete. In contrast, under CO<sub>2</sub> curing condition, concrete prepared with steel slag as the aggregates exhibited higher compressive strengths than those of the concrete cast with natural aggregates, which may be attributed to the enhancement of the interface between the coarse steel slag aggregates and the mortar matrix as a result of carbonation.
- 4 CO<sub>2</sub> treatment on the steel slag concrete improved its volume stability. On the one hand, carbonation directly reduced the contents of periclase and free-CaO in the concrete. On the other hand, the carbonate products layer formed around the coarse steel slag aggregate may play a key role in preventing the water from diffusing into the inner part of steel slag aggregate to further hydrate the free-CaO and periclase, which may lead to excessive expansion. This implies that, with accelerated CO<sub>2</sub> curing, steel slag could be potentially used as fine and coarse aggregates in concrete. However, the long-term volume stability of the steel slag concrete needs further investigation.
- 5 The incorporation of more reactive magnesia compared to quicklime in the concrete produced greater compressive strength after accelerated carbonation.

## Acknowledgements

The authors are grateful to the financial supports from the open foundation of Anhui province key laboratory of advanced building materials of Anhui Jianzhu University (JZCL201601KF), National Natural Science Foundation of China (51461135003, 51502134, 51308004), EPSRC UK (EP/M003159/1), and the Priority Academic Program Development of Jiangsu Higher Education Institutions (PAPD).

## References

- [1] K.L. Scrivener, R.J. Kirkpatrick, Innovation in use and research on cementitious material, *Cem. Concr. Res.* 28 (2008) 128–136.
- [2] S. Monkman, Y. Shao, Integration of carbon sequestration into curing process of precast concrete, *Can. J. Civ. Eng.* 37 (2010) 302–310.
- [3] C. Unluer, A. Al-Tabbaa, Enhancing the carbonation of MgO cement porous blocks through improved curing conditions, *Cem. Concr. Res.* 59 (2014) 55–65.
- [4] P. de Silva, L. Bucea, D.R. Moorehead, V. Sirivivatnanon, Carbonate binders: reaction kinetics, strength and microstructure, *Cem. Concr. Compos* 28 (2006) 613–620.
- [5] J. Chang, Y. Li, M. Cao, Y. Fang, Influence of magnesium hydroxide content and fineness on the carbonation of calcium hydroxide, *Constr. Build. Mater* 55 (2014) 82–88.
- [6] D.K. Panesar, L. Mo, Properties of binary and ternary reactive MgO mortar blends subjected to CO<sub>2</sub> curing, *Cem. Concr. Compos* 38 (2013) 40–49.
- [7] L. Mo, F. Zhang, M. Deng, Effects of carbonation treatment on the properties of hydrated fly ash-MgO-Portland cement blends, *Constr. Build. Mater* 96 (2015) 147–154.
- [8] X. Zhu, H. Hou, X. Huang, Enhance hydration properties of steel slag using grinding aids by mechanochemical effect, *Constr. Build. Mater* 29 (2012) 476–481.
- [9] A. Mahmoud, B. Ali, Laboratory studies to investigate the properties of CIR mixes containing steel slag as a substitute for virgin aggregates, *Constr. Build. Mater* 26 (2012) 475–480.
- [10] N.H. Roslan, M. Ismail, Z. Abdul-Majid, S. Ghoreishiamiri, B. Muhammad, Performance of steel slag and steel sludge in concrete, *Constr. Build. Mater* 104 (2016) 16–24.
- [11] S.I. Abu-Eishah, A.S. El-Dieb, M.S. Bedir, Performance of concrete mixtures made with electric arc furnace (EAF) steel slag aggregate produced in the Arabian Gulf region, *Constr. Build. Mater* 34 (2012) 249–256.
- [12] E. Anastasiou, F.K. Georgiadis, M. Stefanidou, Utilization of fine recycled aggregate in concrete with fly ash and steel slag, *Constr. Build. Mater* 50 (2014) 154–161.
- [13] L. Muhmood, S. Vitta, D. Venkateswaran, Cementitious and pozzolanic behavior of electric arc furnace steel slags, *Cem. Concr. Res.* 39 (2009) 102–109.
- [14] S. Kourounis, S. Tsivilis, P.E. Tsakiridis, G.D. Papadimitriou, Z. Tsibouki, Properties and hydration of blended cements with steelmaking slag, *Cem. Concr. Res.* 37 (2007) 815–822.
- [15] Z. Ghoulleh, R.L.L. Guthrie, Y. Shao, High-strength KOBM steel slag binder activated by carbonation, *Constr. Build. Mater* 99 (2015) 175–183.
- [16] M. Mahoutian, Z. Ghoulleh, Y. Shao, Carbon dioxide activated ladle slag binder, *Constr. Build. Mater* 66 (2014) 214–221.
- [17] L. Mo, F. Zhang, M. Deng, Mechanical performance and microstructure of the calcium carbonate binders produced by carbonating steel slag paste under CO<sub>2</sub> curing, *Cem. Concr. Res.* 88 (2016) 217–226.
- [18] M.A. Boone, P. Nielsen, T. De Kock, M.N. Boone, M. Quaghebeur, V. Cnudde, Monitoring of stainless-steel slag carbonation using X-ray computed microtomography, *Environ. Sci. Technol.* 48 (2014) 674–680.
- [19] Shu-Yuan Pan, Rahul Adhikari, Yi-Hung Chen, Ping Li, Pen-Chi Chiang, Integrated and innovative steel slag utilization for iron reclamation, green material production and CO<sub>2</sub> fixation via accelerated carbonation, *J. Clean. Prod.* 137 (2016) 617–631.
- [20] M. Bodor, R.M. Santos, G. Cristea, M. Salman, Ö. Cizer, R.I. Iacobescu, Y.W. Chiang, K. van Balen, M. Vlad, T. van Gerven, Laboratory investigation of carbonated BOF slag used as partial replacement of natural aggregate in cement mortars, *Cem. Concr. Compos* 65 (2016) 55–66.